Chapter 1

General introduction

1 History of food fraud

The longing for economic success has tempted food sellers to maximize their profit margins ever since. Unfortunately, there have always been people trying to gain their profits using dishonorable strategies. Thus, food fraud is a widespread phenomenon since the beginning of food commerce. For example, in Ancient Rome, fresh food was mixed with spoilt food, expensive products were blended with inferior goods, and coloring or flavoring additives were introduced to mask impurities (Schieber, 2008). The first food frauds entailed the first regulating processes as well. In Ancient Rome and Athens, laws considering the adulteration of wine with flavors and colors were enacted (Sumar & Ismail, 1995). In medieval times, France and Germany passed food control statutes, King John in England made a decree regarding penalties for the adulteration of bread, and extensive laws were enacted by Henry III regarding the adulteration of human food. For centuries, food adulteration was common, but suitable analytical methods were missing to disclose such practice. In consequence, consumers were largely unaware of the extent of food fraud they were exposed to. In the 19th century, the German chemist Frederick Carl Accum was one of the first who elucidated the practice with his most famous book *A Treatise on Adulterations of Food and Culinary Poisons* published in 1820 (Shears, 2010). He availed the principle of "naming and shaming" by naming individual merchants, their offence and the resulting penalty. Apart from this, he described methods for detecting adulteration and can therefore be considered as one of the pioneers of food authentication. Dr. John Postgate and Arthur Hill Hassall have expedited the awareness that systems for the detection and monitoring of food adulteration were necessary. After a comprehensive investigation of foods in the British food trade, the first Food Adulteration Act was passed in 1860 and revised in 1872. The demand of food inspectors came up and in 1874 the Society of Public Analysts was founded with Hassall as its first president. Since then, the

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growing knowledge about the composition of food due to advances in analytical chemistry has evolved a competition between adulterators and analysts.

2 Adulterated food today

Today, the manipulation of foodstuff can be categorized into food quality, food safety, food fraud, and food defense. Food quality and food safety reflect unintended adulterations, whereas food fraud and food defense imply intentional actions. Apart from this, a further categorization can be made considering economical aspects or the health risks of manipulation. Risks belonging to the fields of food quality and food fraud generally underlie financial aspects such as the motivation of gaining higher economic profits. Thus, a food quality risk would be the accidental bruising of a fruit which occurs due to mishandling caused, for example, by an inappropriate cost reduction in the production process. On the other hand, the addition of melamine to milk in order to pretend a high protein content with the motivation to increase the margin is considered a food fraud. In contrast to these economically motivated actions, harmful food alterations are considered in food safety and food defense. Food safety covers, for example, the unintended contamination of raw vegetables with pathogenic microorganisms due to poor protection and control during the production process. A risk of food defense is reflected by the intentional contamination of food with toxic substances with a revenge intent against a store or manager by injuring consumers (Spink & Moyer, 2011).

In order to protect consumers from all these types of food manipulation, various actions of prevention and controlling are enforced. In Germany, for example, in the first place, food business operators are responsible for the safety of the foodstuff they distribute. Via self-monitoring and quality management systems they have to ensure the compliance of established laws. This includes, among others, the application of a HACCP concept to ensure safe processing and production of foodstuff as well as the traceability of every product on the basis of labeling and documentation. This embodies the first step of food control which minimizes the occurrence of unintended food contaminations and makes a quick identification of the source of occurring problems possible. The official food control conducts the second step by a supervision of the control accomplished by the economy. Through randomized and risk-oriented controls of foodstuff and food production sites, it is intended to protect the consumers' health and information. All levels from growers and breeders via processors to vendors are covered by about one million factory inspections and approximately 400,000 analyzed food samples in Germany per year. In case of an offense of established food law, the authorities are

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enforced to apply various sanctions from cautioning via penalty charge to criminal process. If foods are hazardous to health, they are not allowed to be placed on the market or have to be removed thereof.

According to an EU report, the current “top ten” products at risk of food fraud are olive oil, fish, organic foods, milk, grains, honey and maple syrup, coffee and tea, spices, wine and fruit juices (de Lange, 2013). Olive oil, for example, has superior organoleptic properties and health benefits as compared to other oils. Blending with cheaper seed oils, refined olive oil or mildly deodorized olive oils in order to draw the high profit of extra virgin olive oil are considered the main adulteration problems (Wójcicki et al., 2015). Fish underlies wrong labeling in terms of species, geographical origin or production (European Commission, 2015; Upton, 2015). Similar labeling offenses occur for organic foods if they contain conventional food products (Capuano et al., 2013). In terms of milk, valuable compounds such as milk fat are often extracted or the milk is diluted, which is often masked by the addition of various adulterants (Handford et al., 2016). Honey is controlled in regard to the compliance of sugar content and labeling the correct botanical source and geographic origin (European Commission, 2015). Coffee can show an inappropriate quality of the beans considering species, geographical origin and defective beans. Apart from this, other substances (coffee husks and stems, maize, barley, chicory, wheat middlings, brown sugar, soybean, rye, triticale, and açaí) have been added to coffee blends in order to increase profit margins (Toci et al., 2016). Tea is also diluted via the addition of foreign plant materials or inorganic matter (Dhiman & Singh, 2003), whereas spices even show harmful additives such as sudan red or other red dyes (Moore et al., 2012). Wine and fruit juices underlie the dilution with water and the addition of cheap ingredients to mask adulteration. In addition, high prized fruits are sometimes blended with cheaper species (Widmer et al., 1992; Zhang et al., 2009; Nuncio Jauregui et al., 2014). In conclusion, the main food product adulterations occur because of the dilution with inferior additives and false labeling in terms of species, geographical origin, and way of production. In order to avoid an unfair competition and ensure consumer protection, there has to be the possibility to detect these adulterations. This problem is addressed by the analytical discipline of food authentication.

3 Food authentication

One of the most important processes of food control and quality control is the detection of fraudulent practice. Food authentication fulfills this demand as its purpose is the verification of the compliance of foodstuff with the specifications of the products. Reviewed features are,

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among others, the origin (species, geographical or genetic), the production method (conventional, organic, traditional procedures, free range) or the processing technologies applied (irradiation, freezing, microwave heating, enzymatic treatment) (Danezis et al., 2016). In the context of globalized food markets, which lead to an increasing product variability and availability, consumers demand reliable knowledge about the origin and composition of the food products they eat and drink. Producers are also interested in authentic commodities and food preparations used for high-value products as they have to fulfill national legislation, international standards and guidelines. In the course of expanding food markets, the scientific interest in the development of food authentication increases, too. The number of published studies on food authentication increased exponentially in the past 10 years from 409 articles between 2006 and 2008 to 907 for the period from 2012 to 2014 (Danezis et al., 2016). Its great importance is also reflected by numerous research projects funded by the German Federal Ministry for Economic Affairs and Energy and representatives of industries. The present dissertation has also arisen from such a project dealing with fruits and fruit juices, which are part of the "top ten" counterfeit products as mentioned above. It was the IGF research project 16645 N laying the foundation of the present work. Its assignment was the examination of proper authenticity markers and quality attributes for various fruits. For this purpose, more than 200 authentic fruit samples and fruit juice samples from all over the world were acquired in order to establish a valid database. On that account, the polyphenol profiles of the *Vaccinium* L. species cranberry, bilberry, and lingonberry as well as of pomegranate (*Punica granatum* L.) were investigated, which enabled a species authentication. Moreover, the geographical origin was authenticated by the analysis of stable isotopes. The ongoing topicality of food authentication is also reflected by the initiation of further projects. In 2016, a comprehensive project called "FoodAuthent" funded by the German Federal Ministry of Food and Agriculture was launched. A collaboration of the German Federal Ministry of Risk Assessment, IT specialists, and further analytical institutes aims at the development of a system for the acquisition, analysis, and utilization of food authenticity data. The modern approach of fingerprinting analysis shall deliver a database of characteristic fingerprints for various foodstuffs, which will be accessible via a cloud computing system (BfR, 2016).

4 Modern techniques detecting food fraud

There are multiple ways of testing food authenticity reflected by numerous analytical techniques and principally differing approaches. The classical authenticity assessment of food

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usually follows the principle of a targeted analysis. This “bottom-up” approach implies that specific marker compounds are analyzed, which were previously defined. Subsequently, the results are evaluated on the basis of a univariate data analysis, which verifies the presence or absence of marker compounds and the compliance or violation of established limits. Combined with a great effort to validate the analytical method, the targeted analysis provides a reliable authenticity assessment. The drawback is the fact that only those adulterations can be detected which are covered by the defined target compounds. Any fraudulent practice affecting so far unconsidered compounds would remain undisclosed.

On that account, the non-targeted approach found its way into the field of food control. It derives from the principle of “metabolomics”, which describes the scientific study of small molecules, the metabolites, of a biological system. Based on a comprehensive chemical analysis, it is the aim to detect as many substances as possible (Roessner et al., 2011). After gaining popularity in the fields of pharmacy and toxicology, this “top-down” approach has also gained importance in food and feed science in recent years. The great amount of data obtained thereby has to be evaluated on the basis of a multivariate data analysis in order to receive comprehensible results. Often applied multivariate models are hierarchical cluster analysis (HCA), principal component analysis (PCA), linear discriminant analysis (LDA), and partial least squares (PLS) (Riedl et al., 2015).

The application of non-targeted analysis in food science can be categorized into the two groups of food profiling and food fingerprinting, similar to the distinction made in metabolomics (Koek et al., 2011). Food profiling reflects a hypothesis-driven approach concentrating on a limited number of compounds or a compound class. After a non-targeted analysis, compounds have to be identified and can be quantified in order to build a compound database to perform profile comparison during the authentication of further samples. For the description of the mentioned profile, usually multivariate data analyses have to be used. Advantages are a high sensitivity and selectivity due to compound specific sample preparations. Food fingerprinting renounces the identification and quantification of individual compounds and reflects a high throughput screening. If any sample preparation is applied, it is usually conducted in a simple way and all compounds, or as many as possible, are detected. Thereby, unexpected additives or deviations can be revealed. This design allows the investigation of multiple objectives in one analysis run. Yet, a good sample database is necessary for authentication based on multivariate modeling. A comprehensive overview of the non-targeted approaches is given by Esslinger et al. (Esslinger et al., 2014).

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